BSE-504: LEACHATE TREATMENT USING LIMESTONE FILLED-TANK AND CONSTRUCTED WETLAND

***Nadiah bt Mokhtar, Noor Suraya bt Romali, Siti Fatimah bt Che Osmi, Norzamzila bt Mustafa, Muhammad Azizuddin b Mohd Haris, Noor Shida bt Mohd Noor**

Faculty of Civil Engineering and Earth Resources, Universiti Malaysia Pahang,
Lebuhraya Tun Razak, 26300 Gambang, Kuantan, Pahang, Malaysia.
E-mail: nm.nadiah@gmail.com

Abstract

The suitability of eco-limestone filter, consist of limestone filter and constructed wetland (CW) to treat the landfill leachate were investigated. The systems offer an eco-engineering leachate treatment using limestone filter (abiotic component) and wetland (biotic component). A continuous flow mode study has been used to treat raw leachate from Kuantan Landfill. In this study, limestone sized of 14mm, 10mm and 5mm with the ratio of 10:20:70 were used as a filter media in limestone-filled tank. Two types of CW have been applied; sub-surface flow (SSF) system and free water surface (FWS) system. The treated leachate was analyzed for pH, BOD, COD and heavy metal such as Pb, Cu, Mn, Zn and Cr. The results demonstrate that eco-limestone filter was effective to treat leachate especially by using SSF system. The pH of leachate wastewater before and after treatment was complying with standard for discharge leachate as stated in Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009 published by Department of Environment (DOE**)**. For BOD_5, the percentage removal was gradually increased with the increase of contact time. The removal was recorded up to 56% and 50% using SSF and FWS, respectively. The COD, Ammoniacal Nitrogen (A-N) and TSS were removed up to 97%-99% for both systems. Meanwhile, for heavy metals, both systems were capable to attenuate heavy metal and removed it from leachate up to 100%. The system has been run for 10 days contact time, with the flowrate, Q= 4L/min.

Key word: *Adsorbent, Continuous flow-mode, Free Water Surface System, Heavy Metal, Sub-surface Flow System,*

INTRODUCTION

Presently, the amount of solid waste produced in Kuantan is about 500 tons daily, consisting of 60% domestic waste and 40% of industrial and construction waste (Mohd Shahir Zahari *et al*., 2010) which contribute to large volume of leachate generated. For tropical countries like Malaysia, problem arise from leachate production from solid waste generation is a major concern as leachate being generated at large volume especially during rainy season each year. Due to that, treatment of landfill leachate is of concern because it has the potential to degrade the environment. Leachate can be classified as a potential hazardous waste from landfill sites hence needs further treatment.

Leachate is defined as liquid that has percolated through solid waste and has extracted dissolved or suspended materials from it. It arises from the biochemical and physical breakdown of waste (Lu *et al*., 1985). The composition of leachate depends on a variety of parameters, such as the type of waste, site hydrology, landfill type, landfill operation and landfill age (Foul *et al*., 2009). Leachate initially is a high-strength wastewater, contains high concentration of organic matter, inorganic matter and heavy metals (Qasim and Chiang, 1994). The risks from waste leachate are due to its high concentrations of heavy metals. Heavy metal such as Cr, Ni, Zn, Fe, Cr, Cu and Pb could cause serious water pollution and threaten environmental (Foul, *et al*., 2009). Unlike organic pollutants, heavy metals do not decay and thus pose a different kind of challenges for remediation. Because of it toxicity, the presence of heavy metals in excessive quantities will interfere with many beneficial uses of the water. (Aziz *et al*. 2007)

Leachate treatment is very complicated, expensive and often requires multiple processes as reported by Ozturk and Bektas, (2004). Many treatment processes were tested and operational ranges and performance levels were established. Several technologies such as oxidation, sedimentation, ion exchange, membrane filtration, chemical precicpitation, reverse osmosis, air stripping and adsorption have been applied for leachate treatment (Foul *et al.*, 2009).

Previous study investigated by multiple researchers has found that limestone is an effective natural geological material for the treatment of water contaminated by heavy metals. The use of limestone for removing metals from water and industrial wastewater was found to be effective. More than 80% of heavy metals such as Copper, Iron, Manganese, Cadmium and others can be removed using a batch or continuous flow filtration process (Aziz *et. al*., 2004)..

Limestone can increase the effectiveness of wetland treatment systems compatible with the objectives of the project. The systems offer an eco-engineering leachate treatment using limestone filter (abiotic component) and wetland(biotic component). As ecology consist of both factor that interrelated and function in an orderly manner, this system promote a finite system to be used as alternative leachate treatment in green technology. The results of the study demonstrate an ecotechnological means to treat leachate that is high in heavy metal.

The benefit by using this method of treatment is it decreased energy consumption by using natural processes rather than conventional, efficiently removed many pollutants from wastewater and also can enhance the environment by providing a habitat for vegetation, fish and other wildlife and lowered construction, transportation and operation costs. In this regard the present study has been undertaken to (1) investigate the effectiveness of limestone filter and CW on the treatment of raw leachate, and (2) determine the effect of contact time and treatment method to retain pollutant from raw leachate In this study, leachate is generated from Kuantan Jabor-Jerangau landfill site (Kuantan Landfill) which situated at Kuantan, Malaysia.

METHODOLOGY

Leachate Characterisation

Raw leachate sample was collected from Kuantan Landfill, Pahang. The samples were collected from the influent of the detention pond in 25*l* polyethylene container. Upon collection, the leachate was preserved at 4°C in accordance with the Standard Methods for the Examination of Water and Wastewater (APHA, AWWA, WEF, 1992). The quality of the initial raw leachate taken from the Landfill is summarized as per Table 1. Standard used for this experiment was Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009 published by Department of Environment (DOE) which shows acceptable conditions for discharge of leachate to the environment.

Table 1: Characteristics of raw leachate from detention pond at Kuantan Landfill

Parameter	Unit	*Standard	**Initial Reading of Raw Leachate
BOD_5	mg/L	20.0	38.98
COD	mg/L	400.0	6470.0
Ammoniacal Nitrogen	-	5.0	181.0
Total Suspended Solid	mg/L	50.0	513.0
Iron (Fe)	mg/L	5.0	6.0
Lead (Pb)	mg/L	0.1	12.0
Copper (Cu)	mg/L	0.2	9.0
Manganese (Mn)	mg/L	0.2	4.8
Zinc (Zn)	mg/L	2.0	8.4
Chromium (Cr)	mg/L	0.2	0.5

* Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009
**Raw Leachate for the period of April2012

In general, all parameters considered for the experiment exceeded the standard for discharge limit as prescribed by Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009. The low BOD/COD ratio (0.05) indicates that the leachate was stable and difficult to be further degraded biologically (Jokela *at al*., 2002). The concentration of Ammoniacal Nitrogen was very high, that was believed reflecting by the release of soluble nitrogen from solid waste as has been reported by Jokela et al., 2002.

Limestone

The limestone was obtained from Kuari Tinjau Makmur Sdn. Bhd. located at Felda Bukit Sagu 4, Kuantan, Pahang. The limestone size used as a filter media was 14mm, 10mm and 5mm which mixed using a ratio of 10:20:70, respectively. The limestone was separated using sieve analysis. Table 2 below shows the physical properties of limestone:

Table 2: Physical properties of the limestone

Physical properties	Unit
Particle size	30-50mm
Porosity	0.5
Bulk density	15,100 kg/m^3

Constructed Wetland Plant

A common *Typha latifolia* and *Echornia Crassipes* has been chosen as CW plant as it is widely available in Asia region. The plant was obtained from Universiti Malaysia Pahang, Gambang campus area.

Experimental Setup

The experiments were conducted by using reactor system consist of balancing tank, limestone filter and CW. The system was of plug-flow type and the leachate was flow by gravity force. The limestone filter was made of acrylic material with dimensions of 1.5m length × 0.35m diameter that direct connected to two (2) vegetation cells size of 1m length x 0.5 m wide x 0.6 m depth. The 14mm, 10mm and 5m size of limestone were filled in the filter tank, while the *Typha latifolia* (for SSF system) and *Echornia Crassipes* (for FWS system) were planted in the vegetation cells. Figure 1 illustrated the schematic diagram of the continuous flow mode study setup. The experiment was conducted under natural environment conditions which will be exposed to sunlight and open area.

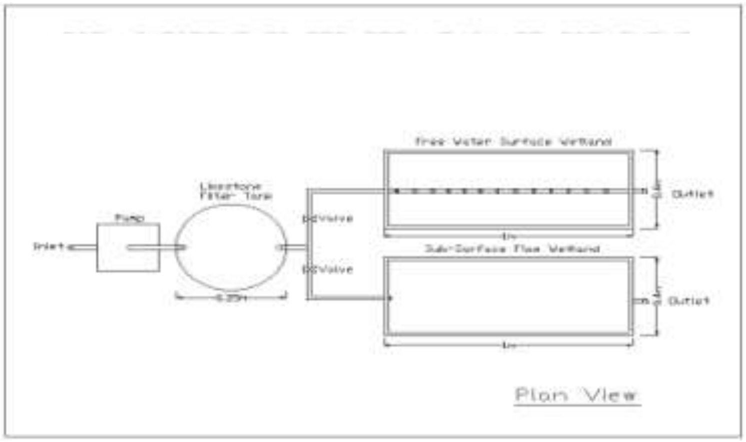


Figure. 1: Schematic diagram of the continuous flow mode study

This experimental work was carried out for 10 days. A volume of 50-L of leachate sample was put in the balancing tank prior to the experimental run. Every 24 hours, the effluent at the outlet of both CW were collected and analyzed. Water samples of 10mL were collected from both systems and stored in the freezer immediately to slow the oxidation reaction and were analyzed for Fe, Pb, Cu, Mn, Zn and Cr by using Gas Chromatography. Spectrophotometer (Model Hach DR/2500) was used to measure Ammoniacal Nitrogen. The pH of the samples was also measured using a pH meter (Model 805MP). Meanwhile TSS was analyzed using Standard Method for the Examination of Water and Wastewater (APHA 2540 D).

RESULTS & ANALYSIS

Based on the results obtained from the experimental procedure, it can be observed that eco-limestone filter for both method (SSF and FWS) have significant effect in adjusting wastewater quality. Table 3 indicates the performance of the filter system in treating leachate at 10 days duration time.

Table 3: The performance of eco-limestone filter in treating raw leachate within 10 days

Parameter	**Unit**	**Standard**	**Initial Reading of Raw Leachate**	**FWS**	**SSF**
pH	-	6-9	7.68	7.64	7.67
BOD_5	mg/L	20.0	38.98	19.432	17.0
COD	mg/L	400.0	6470.0	185	147.0
Ammoniacal Nitrogen	-	5.0	181.0	10	1.5
Total Suspended Solid	mg/L	50.0	513.0	4.300	5.0
Iron (Fe)	mg/L	5.0	6.0	0.001	0
Lead (Pb)	mg/L	0.1	12.0	0.008	0
Copper (Cu)	mg/L	0.2	9.0	0	0
Manganese (Mn)	mg/L	0.2	4.8	0	0
Zinc (Zn)	mg/L	2.0	8.4	0	0
Chromium (Cr)	mg/L	0.2	0.5	0	0

After 10 days contact time, both treatment systems were successfully treating raw leachate to meet the standard limit.

Biochemical Oxygen Demand (BOD)

Figure 2 below is referred. The initial result for 5-day biochemical oxygen demand (BOD_5) of leachate sample was 38.98 mg/l that was exceeding the standard of discharge leachate. As refer to Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009 , the acceptable condition of BOD_5 for discharge leachate is 20mg/l.

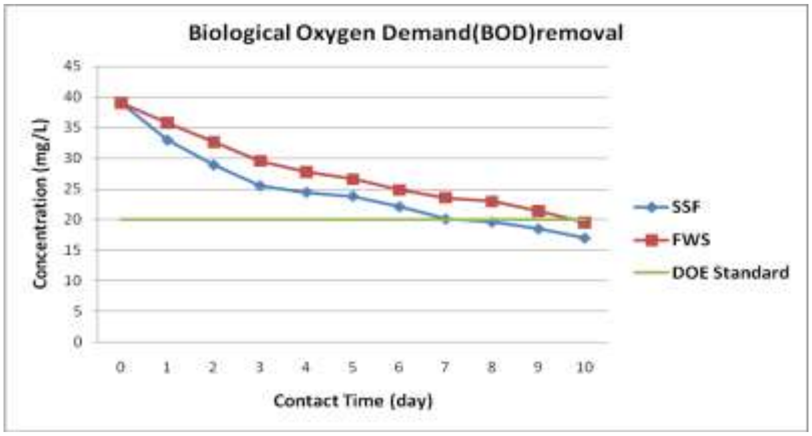


Figure. 2: Effect of eco-limestone filter (SSF & FWS) on BOD concentration (mg/L)

Figure 2 also verifies that there was a significant removal of BOD on both treatment method. There is no exact journal stated that limestone can remove BOD. However, Mukherjee (1968) pointed out that when the influent pH is greater than 7.2, increasing detention period will increases the BOD removal. Limestone in filter could promote the attached aerobic biofilm yielding high performance of organic removal. Meanwhile Sartaj *et al*., 1999 reported that wetland systems can significantly reduce BOD.

Within 10 days contact time, the removal was recorded to achieve 56.4% and 50.15% for SSF and FWS, respectively. The graph depicted that treatment system using SSF was more effective in BOD removal as compared to FWS as SSF took 7 days to meet standard limit for BOD concentration discharge.

Chemical Oxygen Demand (COD)

The initial COD for Kuantan Landfill was recorded high that is 6470.0 mg/l which was exceeding the standard of discharge leachate. According to Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009 , the acceptable condition of COD for discharge leachate is 400mg/*l*.

Figure 3 shows that COD is decrease significantly prior to the treatment at both methods. The COD values continue to decrease significantly with the contact time. Wetland plant can reduce the chemical constituents inside leachate by absorbing through root zone.

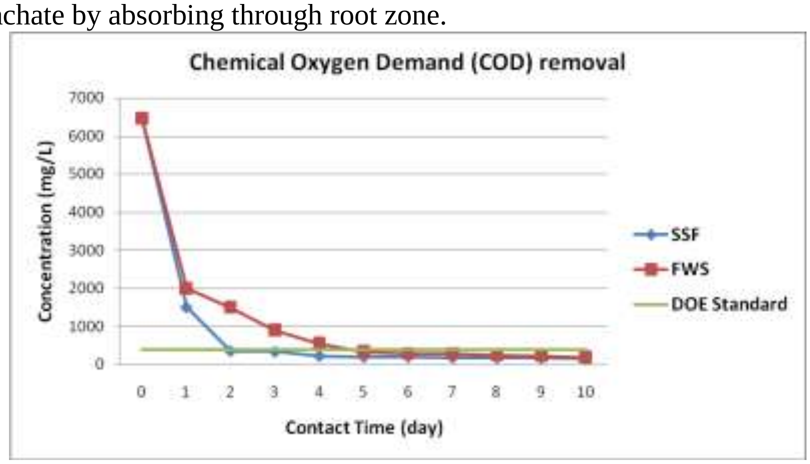


Figure 3: Effect of eco-limestone filter (SSF & FWS) on COD concentration (mg/L)

The removal was recorded to achieve 98% and 97% for SSF and FWS, respectively. The graph depicted that treatment system using SSF is more effective in COD removal as compared to FWS when SSF needs only 2 days contact time to comply with standard limit whereas SSF took 5 days contact time to reach the standard.

Ammoniacal Nitrogen (A-N)

The initial A-N for Kuantan Landfill was recorded high that is 181.0 mg/l while the acceptable condition of TSS should be less than 5mg/*l* as prescribed by Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009.

Figure 4 shows that A-N was decrease significantly prior to the treatment at both methods. The A-N values continue to decrease significantly with the contact time. The increase in the removal of AN with time may be attributed to air stripping phenomena due to increase in pH (Ozturk et al.,2003). Wilai Chiemchaisri et al., 2007 has reported that any soil was effective material in treatment of nitrogen. Water Hycinth plants survived in a pH range of 4.0 to 8.0 and found to enhance the nitrification process (El-Gendy, 2004). Microbial metabolism also affords removal of inorganic nitrogen such as nitrate and ammonium in wetland which would transform nitrate into nitrogen gas.

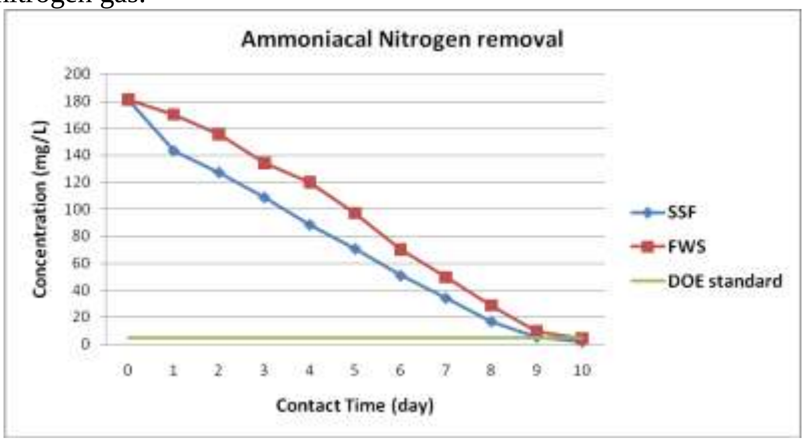


Figure 4: Effect of eco-limestone filter (SSF & FWS) on COD concentration (mg/L)

The graph depicted that both treatment system using SSF and FWS was effectively in AN removal with the removal percentage recorded was 99% and 98%, respectively.

Total Suspended Solid (TSS)

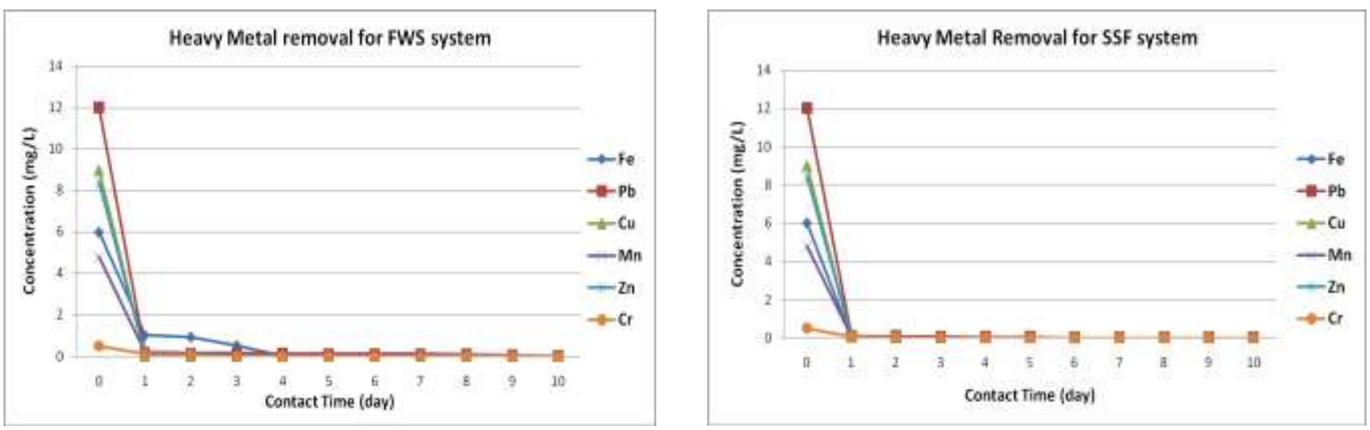
The initial TSS for Kuantan Landfill was recorded high that is 513.0 mg/l while the acceptable condition of TSS should be less than 50mg/*l* as prescribed by Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009.

Figure 4 shows that TSS was decrease significantly prior to the treatment at both methods. The settleable solids are removed easily by sedimentation in CW since wetlands systems generally have long hydraulic retention times. The removal was reported achieved 99% and 98% for SSF and FWS system, respectively. The effluent has met the standard only within 1 day contact time for FWS, and 2 days contact time for SSF.

Figure 4: Effect of eco-limestone filter (SSF & FWS) on TSS concentration (mg/L)

Heavy Metal Removal

Figure 5(a) & (b) depicted the removal efficiency of heavy metal using FWS and SSF system. From the graph, it is observed that the treatment system is capable to attenuate heavy metal from raw leachate. The leachate sample was analyzed for Fe, Pb, Cu, Mn, Zn and Cr Both treatments had totally removed heavy metal from leachate by 100%. The removal of heavy metals was found effective in SSF method where the removal took only 24hours to be removed from the leachate. Meanwhile, Fe concentration in FWS method took 4 days contact time to be removed from leachate.



(a) (b)

Figure 5: (a) Heavy metal removal using (b) Heavy metal removal using FWS system

The Fe concentration in leachate sample was totally removed after 10 days treatment. From Figure 5, 100% of the Fe was removed from the sample only after 1 days in both treatment methods. This has agreed with Ghaly(2007) finding which pointed that the limestone can successfully removed 100% of the Fe from solution on a daily basis. Previous finding by Ghaly et *al*. (2007) also indicates that Fe usually drains from landfills in the reduced ferrous from (Fe^{2+}). At a pH greater than 3.5 with oxygen present, ferrous iron will oxidize to ferric iron as follow:

$$4Fe^{2+} (aq) + O_2(g) + 4H^+(aq) \rightarrow 4Fe^{3+(}aq) + 2H_2O \quad (1)$$

Ferric iron forms iron hydroxide ($Fe(OH)_3$) precipitate as a result of hydroxylation (Fe^{3+} reacting with H_2O molecues) .

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+(aq) \text{ (Acidity) (Ghaly et } al. \text{ (2007)} \quad (2)$$

The surface charge of limestone is also predicted to be a contributing factor for the removal of heavy metals. Divalent metal cation (Fe2+) which is positively charged will be attacted to the negatively charged calcite surface at pH levels higher than 8.3 (Aziz *et al*., 2004).

The Zn concentration in leachate sample after 10 days treatment was 0.04mg/L. This indicates that 82% of the Zn was removed from the sample after 10 days. Sorption of Zn^{2+} ions on calcite in aqueous solution in the presence of humic acids decreased at pH5.5 (Aziz *et al*., 2008).

Statistical Analysis

The significance of the treatment method (SSF and FWS) and contact time was analyzed using Analysis of Variance (ANOVA). Table 4, 5, 6 and 7 summarizes the results of the statistical analysis for the parameter of BOD, COD, TSS and A-N. They support the significance of both variables in pollutant removal at 95% confidence level. Based on the p-value, the treatment method has greater influence in pollutant removal as compared to the contact time within the experimental conditions of the study.

Table 4: Analysis of Variance of BOD removal efficiency

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Treatment Method	8.535335	1	8.535335	35.60603	0.000336	5.317655
Contact Time	28.95859	8	3.619824	15.10047	0.000447	3.438101
Error	1.917728	8	0.239716			
Total	39.41166	17				

Table 5: Analysis of Variance of COD removal efficiency

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Treatment Method	1930613	1	1930613	41.70645	0.000197	5.317655
Contact Time	38213914	8	4776739	103.1905	2.9E-07	3.438101
Error	370324	8	46290.5			
Total	40514851	17				

Table 6: Analysis of Variance of Ammoniacal Nitrogen removal efficiency

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Treatment Method	614.0176	1	614.0176	10.66482	0.011429	5.317655
Contact Time	1132.796	8	141.5995	2.459431	0.112303	3.438101
Error	460.5927	8	57.57409			
Total	2207.407	17				

Table 7 Analysis of Variance of TSS removal efficiency

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Treatment Method	7320.5	1	7320.5	6.749337	0.031719	5.317655
Contact Time	257723	8	32215.38	29.70186	3.64E-05	3.438101
Error	8677	8	1084.625			
Total	273720.5	17				

Mechanism of removal

Limestone

As reported by Aziz *et al*.(2004) limestone gives the highest removal of heavy metal due to two effects. Firstly, the rough surface of limestone itself gives solid contact resulting in chemisorptions of metal ions at low concentrations. Secondly, the presence of dissolve $CaCO_3$ (limestone constituent) had increased the pH which cause metals to precipitate as metal oxide and probably metal carbonate. Previous findings also indicates that apart from precipitation, a small metal quantity is likely retained by ion-exchange with Ca. Aziz *et al(2004)* reported 90% removal of iron by limestone filter from leachate containing 19.5mg/L of iron(Fe). Wajon *et al*.(1985), reported an increases in the removal of iron from contaminated wastewater due to increase in the pH.

Wetland

Mechanism for metals removal in wetlands includes adsorption, chemical precipitation, and plant uptake (Reed *et al*., 1995). Most of the metals uptake by plant through roots and rhizomes . *E.C* can strip Cd from water in a matter of days (Delgado *et al*.,1993). Figure 2(b) shows the removal efficiency between limestone and mix treatment system of limestone and wetland**.**

Figure 2(b) depicted that, treatment system using limestone and *E.C* can enhance the effectiveness of the wastewater treatment. The percentage removal of heavy metal has increase significantly when raw leachate passes through the system**.** The significant removal is clearly stated for Fe and Zn.

CONCLUSIONS

Data from the current study showed that eco-limestone filter using both SSF and FWS systems were highly efficient in the removal of pollutant from raw leachate that contain high in BOD, COD, TSS, A-N and heavy metals (Fe, Pb, Cu, Mn, Zn and Cr). A SSF and FWS planted with *Typha latifolia* and *Echornia Crassipes* was implemented after a limestone filled-tank to enhance the removal of pollutant. In 10 days contact time, both treatment systems were found effective for BOD removal up to 50%-56% for FWS and SSF, respectively. Whereas, 97% to 99% COD, TSS and A-N were successfully removed from raw leachate. The systems were highly efficient to remove heavy metal to meet the standard for discharge leachate. Heavy metals were totally removed from raw leachate in FWS system in 1 day duration, while 4 days duration were taken for SSF to attenuate Fe from the leachate sample. During the experiment, the pH of the wastewater did not exceed 10 thus the wetland ecosystem should be able to adjust to wastewater having a slightly higher pH without suffering adverse effect. The initial cost of constructed wetlands is considerably lower than conventional treatment (Kadlec and Knight, 1996). Therefore, combination of wetland and limestone in the system offer a very effective yet low cost wastewater treatment. For a small community with limited funds, this treatment system is an attractive option to treat leachate from landfill or transfer station. The statistical analysis suggests that treatment method has greater influence in pollutant removal as compared to the contact time within the experimental conditions of the study. As refer to the results, eco-limestone fiter with SSF system has offer a greater removal on raw leachate as compared to FWS system.

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